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THE PLUTONIUM AEROSOL MONITORING PROGRAM AT ANL-IDAHO FACILITIES

by

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Idaho Division

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INTRODUCTION

Argonne National Laboratory is engaged in an extensive research and development program involving plutonium as a fuel in fast breeder power reactors. The ANL-Idaho Facilities have been an integral part of this program since its inception. Plutonium has been used as fuel in the ZPR-III and is the primary fissile component of the EBR-I, Mark IV core. Future plans for the use of plutonium at ANL-Idaho will include the EBR-II Complex, FARET, and ZPPR.

An informal study and information program was initiated within the Idaho Division Health Physics section in 1958 as a result of a series of requests from ANL-Idaho staff personnel for data and information on the detection and monitoring of plutonium aerosols under conditions of reactor operations. The material assembled in the course of the program was originally written in the form of a series of information memoranda circulated within the Idaho Division. These memoranda have been revised, augmented, and updated to form the main body of this report.

The primary topics of discussion are certain of the physical and radiation characteristics of plutonium aerosols (see Glossary, Appendix D), some factors affecting the interpretation of plutonium aerosol samples, and the results of investigations by the Idaho Division Health Physics section of a number of detecting and sampling devices and techniques as applied to plutonium aerosols. The calculated data concerning plutonium are based on material which has not been irradiated and does not contain significant quantities of fission products.

The discussion is deliberately slanted toward monitoring and sampling applications in the vicinity of operating nuclear reactors, since these are the conditions prevailing at ANL-Idaho.

In the interest of brevity, laboratory counting equipment and portable survey instrumentation, and the related techniques of their use, are not discussed. A parallel study program of these subjects has been in progress at ANL-Idaho since 1958. The biological aspects of the inhalation or ingestion of plutonium are not discussed, since adequate treatments may be found in the literature. The treatment assumes familiarity with the basic principles of air sampling, counting, and monitoring devices.

I. PLUTONIUM AEROSOLS

A. Isotopic Composition of Plutonium Reactor Fuel

Plutonium is a mixture of Pu^{239}, Pu^{240}, Pu^{241}, and Pu^{242}, with traces of Pu^{238}. The isotopes of higher mass number are formed by continued neutron irradiation of Pu^{239} after completion of the initial conversion of U^{238} to Pu^{239}. The relative distribution of the isotopes in a given mixture is largely determined by irradiation history. Analysis of one possible mixture is as follows:(1)

Pu^{239}	93.57%
Pu^{240}	5.95%
Pu^{241}	0.465%
Pu^{242}	0.0167%

The specific activity of plutonium, like the relative distribution of isotopes, is dependent on irradiation history. The specific activity of the material above was computed to be 1.58×10^{11} dis/min-g (alpha emitters only.)(2) The isotopic analyses and calculated specific activities of several samples of plutonium, of varying irradiation history, are listed in Appendix A. It should be noted that all typical forms of plutonium emit radiations other than alpha particles and that calculations including other radiations result in values for specific activity different from those given here. The calculations were restricted to alpha emanations only because plutonium emits most of its radiant energy as alpha particles.

B. Characteristics of Plutonium Aerosols

The literature contains a limited amount of information on the physical characteristics of plutonium aerosols. Much of the material is based on experimental work with gram quantities of unalloyed metallic plutonium of unspecified composition. The data are not wholly applicable to ANL-Idaho operations, which are concerned with alloys of varying dilution. Some of the more pertinent data are presented here without comment.

1. Oxidation of Metallic Plutonium

Massive metallic plutonium oxidizes rapidly in moist air, very slowly in dry air. Oxidation processes form an oxide film which is not fully adherent and tends to flake off in small particles that are easily airborne in radiologically significant quantities. Finely divided material, such as lathe turnings, chips, and filings, oxidize more rapidly or may be pyrophoric, the

degree or rate depending on previous surface oxidation conditions, relative surface-to-volume ratio, and humidity conditions.

A fire in a mass of metallic plutonium will cause some of the resulting oxide to become airborne, but the proportion of the original mass becoming airborne is quite small, i.e., 0.0008 to 0.0012%.[3]

No references were found which concerned large quantities of metallic plutonium under conditions of burning or explosion. It is probable that conditions of high temperature and violent dispersion would significantly increase the relative proportion of airborne oxide.

2. Plutonium Aerosol Particle Sizes

Several authors have reported data on the sizes of plutonium aerosol particles. A wide range of observed particle sizes, from 0.004 μ up to approximately 3 μ, has been reported.[3-5] The reported particle sizes are efficiently retained with commonly used air-sampling filter media. In those cases for which the chemical composition of plutonium aerosols has been determined, plutonium dioxide was the only material identified.

3. Radiations from Plutonium Aerosol Samples

When the radiation hazards arising from the use of plutonium are being considered, a distinction must be made between those radiations typical of massive plutonium and of extremely small quantities of plutonium. The total radiation emission from massive material derives from a combination of beta particles, gamma photons, internal-conversion X-rays, spontaneous-fission neutrons, and surface alpha particles. Samples of extremely small quantities of plutonium, such as would be encountered in an aerosol sample, are primarily alpha and beta emitters; radiations other than alpha and beta are not significant for detection purposes when small quantities of material are being considered.

4. Maximum Permissible Concentrations (MPC) in Air for Plutonium Aerosols

National Bureau of Standards Handbook 69 is used as the authority for establishment of the MPC(40) for plutonium aerosols[6] (Note: MPC(40) is ANL nomenclature for Maximum Permissible Concentration, for a 40-hr work week; reference to air is implied). The most pessimistic

value, 2.0×10^{-12} $\mu c/cm^3$, or 4.4 dis/min-m^3, is used. This value is probably conservative. It is based on the assumption that bone is the critical organ and that all plutonium detected is in soluble form, such as the compounds which could be formed in an explosion or fire involving alkali metals. This qualification is necessary because sodium and sodium-potassium alloys are used in ANL-Idaho reactors.

The most probable compound formed by the burning of plutonium is plutonium dioxide, which is an insoluble form. If all plutonium aerosols were insoluble, the MPC(40) could be increased to 4.0×10^{-11} $\mu c/cm^3$, or 88 dis/min-m^3.

5. Magnitude of aerosol Contamination

To illustrate the potential magnitude of the plutonium aerosol problem, consider what might conceivably happen if one gram of plutonium were to be burned in a closed room 20 ft^2 x 10 ft high. Cheever[3] gives a figure of 0.0012% for the amount of plutonium aerosol released from a burning mass of plutonium metal.

Given:

Specific activity: 1.58×10^{11} dis/min-g

Quantity burned: 1 g

Amount of material airborne: 12×10^{-6} g

Calculation:

1. Activity in 12×10^{-6} g (alpha only):

   $(1.58 \times 10^{11}$ dis/min-g$)(12 \times 10^{-6}$ g$) = 1.9 \times 10^6$ dis/min

2. If uniformly dispersed in a room 20 x 20 x 10 ft, or with a total volume of 113 m^3, and assuming that all material released is uniformly suspended in air, the concentration would be

   $$\frac{1.9 \times 10^6 \text{ dis/min}}{113 \text{ m}^3} = 1.7 \times 10^4 \text{ dis/min-m}^3 \quad .$$

3. The MPC(40) for plutonium was given as 4.4 dis/min-m^3. Dividing the previously determined concentration by 4.4 dis/min-m^3 gives the relative MPC(40) value

   $$\frac{1.7 \times 10^4 \text{ dis/min-m}^3}{4.4 \text{ dis/min-m}^3/\text{MPC(40)}} = 3.9 \times 10^3 \text{ MPC(40)} \quad .$$

II. PLUTONIUM AEROSOL SAMPLING AND COUNTING TECHNIQUES

A. Natural Radioactive Aerosols

Analysis of plutonium aerosol samples is complicated by the presence in air, at all times, of a naturally occurring radioactive aerosol of similar characteristics, viz., the gases radon and thoron ($_{86}Rn^{222}$ and $_{86}Rn^{220}$) and their decay products (also called "daughters" or "progeny"). The decay products of radon and thoron are particulates which tend to attach themselves to dust particles in the air and are collected by conventional air-sampling techniques.

1. Sample Evaluation

Radon-thoron daughters are invariably present in the atmosphere in sufficient quantities to mask out the presence of low-level plutonium aerosols when counting is done simultaneously with sampling, or when counting is done immediately, or shortly after, sampling. A procedure adopted by some installations allows about 24 hr of decay time before making a count. This normally permits the radon-thoron daughters to decay to undetectable or insignificant levels and permits the counting of samples for plutonium by simple counting techniques. The disadvantage of this procedure is a 24-hr delay between the end of the sample-collection period and the determination of the amount of plutonium in the sample.

Typical freshly collected radon-thoron daughters have an effective half-life of 30 to 40 min. If several hours are allowed to elapse before counting, the effective half-life is longer, due to the presence of a long half-life thoron daughter (Thorium B, with a 10.6-hr half-life). A 24-hr decay period allows essentially all of the short half-lived material to decay out, and allows the long-lived thoron daughter to pass through two half-lives. Assuming a normal condition, a 24-hr decay period allows the activity due to natural atmospheric radioactivity to drop to 1% or less of the total initial activity present.

Much work has been done on devices or techniques to discriminate between the emanations from plutonium (and other alpha emitters) and those from radon-thoron daughters. Several methods of achieving this discrimination have been examined, and some of these have been adopted for use at ANL-Idaho.

2. Effects of Inversion Conditions

An atmospheric inversion prevents normal mixing of air, with the result that the air below the inversion layer retains almost all of the radon-thoron daughters emitted from the soil. If such a condition exists

for as little as a few hours, the concentration of radon-thoron daughters below the inversion can increase by a factor of ten or more. Where an inversion persists for a longer period of time, i.e., several days, the radon-thoron concentration may increase by a factor of one hundred or more.

Meteorological records for the NRTS show that inversion conditions exist approximately 50% of the time. Inversions are usually nocturnal and dissipate shortly after sunrise. Under some atmospheric conditions, usually occurring in the winter months, continuous inversions have existed for as long as seven days. Such conditions are often associated with large, static, high-pressure areas.

B. Conventional Alpha Air Monitors

The masking effect of radon-thoron daughters on conventional alpha-detecting continuous air monitors rules out the use of these devices as reliable detectors of low-level plutonium aerosols. However, if high levels of plutonium aerosols are a potential hazard, good use can be made of conventional monitors in plutonium operations.

1. The AK-1 Air Monitor

Three locally constructed, continuous alpha-detecting air monitors are being used at ANL-Idaho as "back-up" equipment. The basic unit has been designated AK-1. The collecting surface is a 5 x 12-in. sheet of HV-70 (18-mil) filter paper, backed by an expanded metal screen. This paper is rated at 97% efficiency for 0.3-μ di-octyl phthalate (DOP) particles. Air is drawn through the filter at a rate of 0.25 m^3/min by a vacuum power unit. The detector is a flat probe, 5 x 12 in., with a (1.0 $\pm$ 0.2)-mg/cm^2 aluminized Mylar window. The detector anode is a 10-wire "harp" made from a single strand of 1-mil stainless steel wire looped around Teflon insulator blocks. The detector gas is 90% argon and 10% methane. The detector is mounted directly over the collecting surface at a distance of approximately 0.25 in. and is operated as a proportional counter.

The detector output is fed to a sensitive preamplifier and then to a count-rate meter. Readout is in counts per minute. Efficiency is approximately 30% for plutonium alphas. The system is insensitive to Co^{60} gammas to about 5 r/hr. The monitors are wheel cart-mounted and portable.

2. AK-1 Operational Experience

Two AK-1 units have been in service some eight months. In this period, no major breakdowns have been experienced and maintenance has been minimum. Filters have been changed at irregular intervals. This was

done intentionally to determine filter loadings over long sampling periods, buildup of long-lived activity, etc. Drops in filter air flow of less than 10% in 72 hr have been observed under local operating conditions.

Equilibrium concentrations of radon-thoron daughters have produced count rates varying between the limits of 50 cpm and 2,500 cpm, with the average value estimated at 200 cpm (0.25 m^3/min, 30% efficiency, 30% self-absorption in the collecting media).

3. Efficiency of the AK-1

The AK-1 unit has not been used under actual conditions of major airborne plutonium contamination. Calculations indicate that this unit will respond to a 1 MPC concentration of plutonium aerosol with a buildup of 1 dis(alpha)/min-min of sampling time. If the alarm setting is adjusted to reject alpha activity up to the maximum value observed for radon-thoron daughter products, the required alarm times would not exceed the following values:

Concentration Relative MPC(40)	Maximum Time for Alarm (min)
1 x MPC	4×10^3
100 x MPC	40
1000 x MPC	4

C. Conventional Air Sampling

The continuous air monitors are supplemented by the collection of spot samples using conventional air-sampling techniques and by laboratory analysis of samples taken from the continuous air monitors. In the majority of cases, these samples are counted for both beta-gamma and alpha activities with conventional laboratory counting equipment.

1. Calculation of Data

The results of the sample counts are entered and computed on form IHS-ID-1 (Air Sample Data). Complete counting data and computations are shown in detail on this form to provide a basis for possible future reference or evaluation of the observed results. This form is prepared in duplicate with original copies retained in a permanent file at the originating Health Physics field office and duplicate copies sent to an alternate field office as part of a similar permanent file. All samples which indicate the presence of plutonium-alpha air contamination in excess of 10% of MPC(40) are itemized in the section monthly report.

2. Immediate Counting

It is the policy of the Idaho Division Health Physics section to perform an "immediate" count on all conventional air samples. The purpose is three-fold: first, the results of such counts provide reference data on the magnitude of radon-thoron daughter concentrations; second, there is always a finite probability that an unobserved high-level release of plutonium could have occurred and that the plutonium-alpha concentration deposited on the filter could be significantly higher than the normally observed range of activity for radon-thoron daughters; and third, it is possible, by application of a formula based on the natural ratio (beta-gamma:alpha for radon-thoron daughters) to ascertain with reasonable accuracy whether or not a significant quantity of plutonium is present on a filter, even though the plutonium-alpha activity is "hidden" by a moderate to high concentration of natural radioactivity.(7)

The follow-up procedures on air sample counting are outlined in section operating procedures. A detailed description was not considered pertinent to this report.

D. The Alpha-Beta-Gamma Ratio

An empirical relationship exists between the alpha- and beta-gamma-emitting characteristics of plutonium and of radon-thoron daughters. Radon-thoron daughters emit beta and alpha particles in a ratio of about 2.8 betas to one alpha, whereas plutonium emits an alpha particle coincident with some very weak beta and gamma radiations. If a detector system is sensitive to high-energy beta and gamma radiations and is, at the same time, sensitive to alpha radiations, plutonium will be seen as an essentially pure alpha emitter with a beta-gamma:alpha ratio of the order of 1:10 to 1:100.

In practice, detectors are used which detect and count separately the alpha emanations and the beta-gamma emanations from a sample. The outputs of the "alpha" channel and the "beta-gamma" channel are amplified, and then converted to a count rate meter readout, usually in terms of counts or disintegrations per minute. The outputs of the separate count rate meters are then compared electronically to determine the ratio beta-gamma:alpha.

If the relative efficiencies of the alpha and beta-gamma channels are approximately equal, or if the related count rate meters can be calibrated to equal response, this type of device will give a readout in terms of ratio of beta-gamma to alpha.

If a ratio detection system is used to analyze a sample containing both radon-thoron daughters and plutonium, the observed ratio will fall below that determined for radon-thoron daughters alone, the exact point

being determined by the detection-electronics system characteristics and the relative concentrations of radon-thoron daughters and plutonium. If only a small amount of plutonium is present, the ratio will be shifted slightly downward, but perhaps not enough to be significant. As more plutonium is deposited, the ratio will continue to shift downward until a point is reached which is obviously below the limit of statistical variation for radon-thoron daughters. This is the point at which an adjustable alarm mechanism is set to trigger. The amount of plutonium required to produce this effect, i.e., a drop in the observed ratio beta-gamma:alpha, is a variable dependent on the ambient concentration of radon-thoron daughters.

1. Operational Ratio Monitors

Typical units using ratio monitors collect aerosols by drawing air through a suitably held paper filter of high efficiency (retains 95-97% of 0.3-μ particles of di-octyl phthalate - usually, Hollingsworth and Vose HV-70 or equivalent) and monitor the filtering surface simultaneously with a detector capable of detecting both alpha and beta-gamma radiations, with some manner of provision for separating the detected pulses into appropriate channels.

The Idaho Division has had a limited amount of operational experience with two ratio-type continuous air monitors. One unit was purchased in 1961. The second unit, built to ANL specifications, was borrowed from Argonne's Industrial Hygiene and Safety Division for test purposes in 1961. The units represent two distinct approaches to the ratio detection system.

The first unit is designed to utilize a single counting chamber with a Mylar window and a single collecting electrode to detect both alpha and beta-gamma radiations. Separation is accomplished by a system of pulse-height sorting. The chamber is operated in the limited proportional voltage region. Much difficulty was experienced in the first eight months of operation. A number of electronics adjustments were required on an almost continuous basis to keep the instrument in operation.

It was decided to modify the unit by using dual sampling heads and dual detectors. By use of a discriminating alpha detector on one head and a discriminating beta-gamma detector on the other head, and feeding the respective outputs into appropriate amplifiers, a good separation of pulses was attained. The modified unit has now operated for some ten months. In this period, there were no electronic or mechanical breakdowns, and the ratio system operated in a stable and satisfactory manner.

The modified unit was placed in operational service at EBR-I. Experience at this location indicates that it will perform its function satisfactorily, but that some interpretation of results is necessary because of changes in area background related to reactor operation and variation in observed values for the beta-gamma:alpha ratio.

Environmental conditions relative to reactor operation affect the operating efficiency of the unit. The primary consideration at EBR-I was variation in gamma background incident to reactor operation. In effect, an increase in gamma background, which is proportional to reactor power, increases the apparent ratio seen by the detection system and decreases the sensitivity of the unit. The mode of operation of ANL-Idaho reactors is generally a series of short runs at varying power levels. Changes in alarm level settings during reactor operation are not feasible in what is, theoretically, an unattended automatic monitor. If the alarm level settings are not adjusted, it is apparent that the monitor will be less efficient for plutonium-alpha detection during reactor operation.

Local variations in the natural radon-thoron daughter ratio have been observed; this is a factor for which compensation cannot be made in an automatic monitor because the variations are not predictable.

The total effect of the environmental variables noted above is to reduce the potential effectiveness of the unit to a point below that of the more conventional monitoring devices. Under conditions of normal radon-thoron daughter concentration ($1\text{-}1.5 \times 10^3$ radon dis/min-m^3)(8) and ratio, it was calculated that the ratio monitor described above would respond to a plutonium aerosol concentration of one MPC(40) in 24 hr, or to a concentration of 24 times MPC(40) in one hour. Relatively small variations in radon-thoron daughter concentration and ratio and in ambient gamma background can affect the unit's sensitivity by a factor of ten or more.

The second unit uses the "piggy-back" system of two thin counters, one superposed on the other and both counting a common sample The detector nearest the sample has a thin Mylar window, which passes both alpha and beta-gamma radiations, and is operated as an alpha proportional counter with the output biased to count only alphas. This detector is mechanically separated from the second detector by an aluminum plate sufficiently thick to stop alpha particles, but thin enough to pass energetic beta and essentially all gammas. The second detector is operated on the upper end of the limited proportional voltage region. The output from each detector is amplified separately and fed to the system count rate meter and ratio system.

The unit was used intermittently in the ZPR-III for about six months. It was extremely unstable and required daily maintenance to keep it in operation. It was found that the gamma response during and shortly after reactor operation made operation of the unit impossible. The detector was shielded with lead bricks in an attempt to reduce the response to gamma background from the reactor, but again this was not sufficient to produce satisfactory results. It would have been possible to deactivate

the beta-gamma section and use the monitor as an "alpha-only" monitor, but it was believed that this mode of operation could not be justified under continuous use.

In routine "clean area" operation, where only radon-thoron daughters were present, the ratio response was unstable over extended periods. When reasons for variation of the ratio could be established, the trouble was traced to either the sensitivity or high voltage adjustments. Complete breakdown of the unit could usually be traced to defective electronic tubes. It is possible that many of the observed troubles could have been corrected by appropriate maintenance, which was not available due to insufficient experience with this unit.

2. Ratio Discrepancies

Recent data abstracted from conventional air-sample records at EBR-II and from chart recordings of the ratio monitor at EBR-I indicate some major discrepancies between the locally observed values for the beta-gamma:alpha ratio and those reported at ANL-Illinois.

Normal values of the beta-gamma:alpha ratio are reported to be within the limits 2.0:1 to 3.5:1. ANL-Idaho records showed less than 50% of the data between these limits. The 90% deviation limits for conventional air samples were 1.0:1 to 4.5:1 (with 51 samples reported). Chart records from EBR-I for the sample period indicated ratios varying between the limits of 2.0:1 to 7.0:1, with occasional increases to 10:1. Data recorded for periods of reactor operation are not included in the values given.

The data noted here were originally assembled to corroborate claims for the ratio principle with respect to its application to automatic ratio air monitors. Analysis of the data indicates a number of discrepancies between local results and those obtained and reported elsewhere. If the local results are valid, the claims of stability and reliability for monitors using this principle are open to question.

3. Conclusions

Operational experience with ratio monitors at ANL-Idaho indicates that the use of automatic ratio monitors in a reactor environment should be limited, pending further operational experience with currently available units. In theory, the ratio monitor is best used in a stable environment. The use of ratio monitors at ANL-Illinois has been extremely successful in such applications as radiochemical laboratories and processing facilities. At ANL-Idaho, the variables encountered in a reactor environment reduce the reliability and sensitivity of response of ratio monitors to a point below that of conventional air monitors.

E. Impaction Monitoring

1. Principle

G. W. C. Tait reported a method of separating plutonium aerosols from atmospheric radon-thoron daughter aerosols by the use of an annular impactor to effect a separation by mass and particle size.[9] The annular impactor is a device in which a jet of high-velocity air is caused to make a sharp-angle bend around a curve of very small radius. As the jet carries a dust particle around the bend, the momentum of the particle causes it to trace a path around the bend with a radius which varies in a direct proportion to the size and specific gravity of the particle. Small, low-density particles follow a small radius while larger, high-density particles trace a curve with a greater radius.

It is reported that radon-thoron daughters are almost always associated with small, low-density particles and that plutonium aerosols are composed primarily of somewhat larger and higher density particles.[4,5,8-11] In theory, it is possible to separate radon-thoron daughter aerosols and plutonium aerosols by means of the annular impactor.

Tait reported that if he placed a collecting surface at some small distance from the sharp bend of the air jet, small light particles would pass through the opening while larger, heavier particles collided with or impinged on the collecting surface. By adjustment of the width of the impactor annulus it was possible to obtain a distinct separation of plutonium aerosols from the naturally occurring radon-thoron daughter aerosols. Tait's impactor used a commercial vacuum cleaner motor as a power unit and a small metal disk or planchet as a sample collector. The collection surface was usually coated with a thin film of oil to help in retention of the impacted particles.

Tait's original work was confirmed by a number of independent investigators.[4,10,12] It is noted, however, that certain other investigators disagree with some of the basic assumptions as to plutonium aerosol particle sizes and the calculations pertaining to theoretical efficiency.[3,13] Several investigators have made modifications of Tait's original design, but all their units are basically similar in function. The Health Physics group at Savannah River has done a great deal of work with adaptations of the basic impactor for use in continuous air monitors.[4,12]

2. Operational Impaction Monitors

The Idaho Division, in 1959, built a continuous impactor monitor for plutonium aerosols from blueprints furnished by Savannah River.[4,12,14] The monitor has been in intermittent use at ZPR-III up to the present time.

It uses vaseline-coated Mylar-base film (70 mm, with ASA standard perforations) as an impaction surface, a vacuum cleaner motor as a power unit, and associated drive and detection systems. In operation, a sample is collected for 5 min on one fixed spot on the tape. At the end of the 5-min period, a drive mechanism moves the tape to a position under an alpha-detecting scintillation counter with a simultaneous movement of a fresh section of tape to the sample-collection zone. The sample under the detector is then counted for 5 min while a fresh sample is being deposited at the impaction head. The detector feeds a count-rate meter which reads out on a panel meter and on a graphic recorder. An alarm system is activated by the meter readout (actually an adjustable meter-relay).

This monitor was evaluated by personnel of IHS-ANL.[13] The evaluation report disagrees with the results reported by Lapsley[15] regarding the efficiency of the impaction sampling head for plutonium fume. The basis for disagreement is a difference in reported particle sizes and jet velocity. The results of the evaluation indicate essentially 100% efficiency for plutonium aerosol particles of size greater than 2μ, with efficiency dropping off from 100% at 2μ to zero percent at sub-micron levels.

If a particle size distribution of from 1 to 3μ can be assumed, the device is calculated to have a maximum sensitivity of ten MPC(40) in a 10-min period (5 min of sampling time and 5 min of counting time).

The unit has been in intermittent use at ZPR-III since early 1960. No significant maintenance problems have been encountered.

A second impactor monitor was recently purchased from a commercial vendor. The unit is mechanically similar to the unit described by Collins,[11] which uses 3-in.-wide masking tape as an impaction surface. Drive is continuous at $\frac{1}{4}$ in./min. The detector is an alpha scintillation counter utilizing a ZnS phosphor on a standard photomultiplier. Delay between the sampling head and the detector is approximately 4 min. An adequate evaluation of the unit had not been made prior to preparation of this report.

3. Experimental Data on Impaction Units

D. C. Stevens reports some work done at Harwell with an impactor.[10] He found that radon-thoron daughters were collected with low efficiency (high rejection) and that in most cases the radioactivity collected on the impaction surface was less than 5 dis/min-m^3 (considering radon-thoron daughters only).

Similar work was done at EBR-II with an impactor made in our own shop facility. The operational characteristics of this unit are similar to those reported for the Tait and Stevens units. Results with 85 samples, the majority being atmospheric, taken in the EBR-II area (prior to reactor operation) averaged 19.6 dis/min-m^3 of alpha-emitting radon-thoron daughters. Of the 85 samples, 15 exceeded 30 dis/min-m^3. The highest activity recorded was 210 dis/min-m^3, found on a sample taken on January 9, 1963. It is noted that the period covered by these samples was characterized by local inversion conditions of unusual magnitude, with attendant high concentrations of radon-thoron daughters.

4. Conclusions

An evaluation of the EBR-II data indicates that the unqualified use of impaction devices for plutonium aerosol sampling under conditions of atmospheric inversion may not be justified. If qualifying restrictions must be placed on the use of the impactor (for example, allowing time for decay of radon-thoron daughters), its reported advantages are largely nullified.

III. ANALYSIS TECHNIQUES FOR PLUTONIUM DETERMINATION

A facet of the plutonium-aerosol-monitoring program which must be considered is that under many circumstances plutonium is used in conjunction with uranium and other alpha emitters. In all of the monitoring systems and procedures previously noted, all long-lived alpha activity must be presumed to be emanating from plutonium because these systems are basically incapable of making a clear distinction between uranium and plutonium. In the majority of circumstances, this is not expected to be an insuperable problem. However, conditions could conceivably occur which would be acceptable if the contaminant were uranium, but unacceptable if the contaminant were plutonium.

A. Chemical Separation

It is possible to dissolve the sample-collection media, chemically make a separation of plutonium from uranium, and run a radioactivity analysis for either uranium or plutonium. The method is not suitable for routine operations or when the quantities of material are small. As a further complication, the sampling media best suited for radioactivity-counting procedures are not compatible with chemical separation techniques. This system has a further disadvantage in that the sample is destroyed in the chemical separation process and is not available for subsequent counting.

B. Alpha-particle Pulse-height Analysis

Pulse-height techniques have been applied to plutonium-uranium analysis at the ANL-Idaho facilities, but they are not readily applicable to routine aerosol monitoring because they are complex and time-consuming. However, under some circumstances they may be the only means of information or identification.

The devices noted below are assigned to the ZPR-III physics counting group. Analyses for health physics purposes must be requested from this group. Similar services are provided by the AEC-IDO Analysis Branch.

1. The Frisch Grid Chamber

A device of primary interest as a health physics tool is the "Frisch Grid Chamber." The detector is basically a windowless ion chamber, i.e., the sample is placed inside the sensitive volume of the detector. A screen grid, positively charged, is located below the collector anode to reduce space-charge effects and reduce time-constant considerations. The output pulses from the system are proportional to the number of ion pairs formed. The manufacturer claims 33% efficiency with 2% resolution and negligible background. This instrument is a recent acquisition. In theory, it is capable of resolving the 5.147-Mev and 5.097-Mev alphas from Pu^{239} in the presence of such emanations as the 5.998-, 5.505-, and 7.680-Mev alphas from radon daughters or the 4.195-Mev alpha from U^{238}.

As applied to aerosol samples, the Frisch Grid Chamber is reportedly capable of resolving alpha peaks from samples containing less than 10% of MPC(40) of plutonium alphas.

2. Solid-state Detectors

Solid-state detectors have been used in alpha pulse-height analysis to identify plutonium and other alpha emitters, and some experimental work has been done at ANL-Idaho in an effort to apply this technique to the analysis of air samples. The ZPR-III reactor physics counting group did some preliminary evaluation of a number of solid-state detectors. Their best results were obtained with a unit with a 2.2-cm-diameter effective area. To maintain a minimum resolution of less than 100 kev, it was necessary to position the detector surface 7.0 cm from the sample surface. The detector was very nearly 100% efficient for all alphas reaching the detection surface, but geometric considerations reduced the relative efficiency in this configuration to approximately 6%, with resolution quoted as 78 kev at 5 Mev.

There is no practical limit to the size in which solid-state detectors can be made; however, resolution falls off as size is increased, to the point where large surface detectors become useless as pulse-analysis devices. The physics counting group believes that the 2.2-cm-diameter unit is about as large as can be profitably operated as a pulse-height analyzer, at least in the present state of the art. This device could be satisfactorily used with very "hot" samples, but does not have the sensitivity and resolution of the Frisch Grid Chamber. The unit must be operated under vacuum.

IV. THE ANL-IDAHO MONITORING PROGRAM

The operational plutonium aerosol monitoring program at the ANL-Idaho Facilities is divided into three phases; the coverages provided under each are described below.

A. Continuous Monitoring and Sampling

Continuous monitoring and sampling is provided in all areas where plutonium is present. Minimum coverage is considered to be a continuous "self-monitoring" sampler (either a conventional alpha air monitor or a ratio detection monitor), supplemented by one or more conventional air samplers. Monitoring units are inspected daily for proper operation. All monitors are equipped with chart recorders. All charts are kept as a permanent record. Filters are removed from monitoring units and routinely counted as a double check on the monitor's detection and recording system.

The conventional air-sampling units used for this phase of the program are continuously operating devices sampling at relatively low flow rates. Typical units sample room air at 2 cfm on 2-in.-diameter HV-70 or Millipore filters. Samples are removed daily, Monday through Friday, and counted for alpha and beta-gamma activities.

B. Special Operation Monitoring and Sampling

All operations involving the handling, transfer, examination, or inventory of plutonium are monitored for release of plutonium aerosols by moving one of the continuous monitoring devices to the vicinity of the operation. At other times, special-purpose monitors and samplers may be used. Minimum coverage is considered to be provided by one continuous monitor supplemented by one or more conventional air samplers or special-purpose monitors.

The air samplers used for the purposes noted here may be either low-flow-rate devices as noted in the previous section, or high-volume types. The latter are preferred when a specific operation is of short duration, for then adequate volumes may be sampled in a brief period.

The low-flow-rate samplers utilize carbon-vane vacuum pumps and normally operate at flow rates of 1 to 2 cfm.

The high-volume or high-flow-rate samplers are high-speed metal-vane or rotary positive-displacement devices. Sampling flow rates are dependent on the physical size of the filter, the type of filter, and the power rating of the unit. Depending on the unit and filter used, flow rates are on the order of 8 to 40 cfm.

One example of a special-purpose monitor is an experimental "sniffer." This unit, built in the Idaho Division Health Physics section's instrument shop, consists of a portable alpha survey meter coupled with an air-sampling device to provide a portable unit capable of measuring alpha particles simultaneously with deposition on the paper filter. Air is drawn through the filter at a rate of 10 to 30 cfm. The unit is provided with a "snout" to enable it to probe cavities, openings, and inaccessible areas.

C. Emergency Sampling

If it is known or suspected that a plutonium aerosol release has occurred in an area, either through alarming of the continuous monitor or as a result of a fire, explosion, or other catastrophic incident, high-volume-type air samplers are used to collect samples for analysis. Portable high-volume samplers are located in each area and are maintained in a state of readiness, i.e., filter in place and covered with an easily removable, impervious cap or shield to prevent cross-contamination prior to actual use. Completely portable samplers, with self-contained power supplies, are available on short notice from the AEC-IDO Health Physics monitoring group.

D. Supplemental Procedures

Sample media from conventional air samplers, from the continuous monitors, and from impaction samplers are routinely counted for both alpha and beta-gamma activities. Discriminating counters are used to count for gross alpha and for gross beta-gamma. Alpha counters are calibrated against plutonium standards. Beta-gamma counters are calibrated against Radium D, E, F (combined) standards. The standards used are "secondary" standards, prepared and calibrated by the background counting group of Argonne's Industrial Hygiene and Safety Division. All counters in regular use are calibrated daily for yield or geometry and for residual background.

Long-lived residual alpha activity, i.e., activity remaining after 7-day decay, is assumed to be plutonium, unless positive isotopic or chemical identification is made. Current operating procedure requires isotopic identification on all samples having long-lived residual gross alpha activity greater than 50% of MPC(40) for plutonium after 7 or more days of decay time. Identification is made by pulse-height analysis techniques (noted in Section IIIB). Analyses are available through the ZPR-III Physics Counting Group or through the AEC-IDO Analysis Branch, located at Central Facilities Area at the NRTS.

All gross counts and/or isotopic analyses are entered in the permanent record files of the Health Physics section. All gross counts with long half-life alpha activity exceeding 10% of MPC(40) for plutonium and all positive isotopic analyses for plutonium are itemized in the section monthly report.

Supplementary monitoring of total body plutonium intake is provided by routine urinalysis. This service is provided by the AEC-IDO Analysis Branch at the NRTS.

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APPENDIX A

ISOTOPIC COMPOSITION AND CALCULATED SPECIFIC ACTIVITIES OF PLUTONIUM OBTAINED BY IRRADIATION OF URANIUM-238 [After Steindler[15]]

nvt	Fraction of Total Plutonium					Calculated Specific Activity (dis/min-g)
	Pu^{238}	Pu^{239}	Pu^{240}	Pu^{241}	Pu^{242}	
5×10^{19}	0.17×10^{-6}	0.9939	6.08×10^{-3}	5.19×10^{-5}	0	1.38×10^{11}
2×10^{20}	3.2×10^{-6}	0.9665	0.0323	1.15×10^{-3}	2.18×10^{-5}	1.48×10^{11}
2×10^{21}	136×10^{-6}	0.6917	0.234	0.058	0.016	2.17×10^{11}
1×10^{22}	4340×10^{-6}	0.406	0.262	0.098	0.230	3.56×10^{11}

(Note: The specific activities given above were based on a formula in the Radiological Health Handbook[16] and on the percentage quantities of each isotope present in a given mixture. The unit dis/min-g was used for convenience in applications to air-monitoring and -sampling problems.)

APPENDIX B

DESCRIPTION OF PLUTONIUM-FUELED REACTORS AT ANL-IDAHO

A. ZPR-III

1. Reactor. The fast zero-power reactor experiment (ZPR-III) is a device to obtain neutron physics information necessary for the design of fast power breeder reactors. ZPR-III is a flexible research tool and may be used with a wide variety of fuels. Current research projects at ZPR-III are concerned with plutonium.

2. Fuel. The ZPR-III plutonium fuel is in the form of thin flat plates of a Pu-1.1 w/o Al alloy. The plates are nickel-coated and are sealed in a welded, stainless steel jacket. The ZPR-III fuel is assembled in metal drawers in which plates of fuel are inserted together with plates or slugs of other materials to simulate reactor core configurations. Inasmuch as ZPR-III operates at essentially zero power, buildup of fission products in the fuel is not significant under normal operating conditions.

Operational procedures at ZPR-III require frequent manual handling of the fuel and frequent transfers of the fuel from storage to fuel assemblies and back to storage. All of these operations are conducted in air and without shielding. Certain of these operations are conducted within hoods and others within areas of personnel occupancy. The frequency of potential exposure is high. All procedures in which plutonium fuel is used are monitored constantly by techniques which are intentionally redundant.

B. The EBR-I, Mark IV Core

1. Reactor. The Experimental Breeder Reactor I is a fast, unmoderated, power breeder reactor operating at a nominal power level of 1 Mwt. Heat produced in the core is removed by a NaK alloy coolant and may be dissipated to the atmosphere or utilized to produce steam, which may in turn be used to drive a steam turbine-generator for power production.

2. Fuel. The EBR-I, Mark IV plutonium fuel is in the form of cylindrical slugs of a Pu-10 a/o Al alloy. A typical basic fuel rod is composed of a Zircaloy-2 tubular jacket filled with six slugs, of which the upper and lower are of depeleted uranium. The center core section is composed of four Pu-Al alloy slugs. The jacket is filled with NaK for thermal bonding.(17)

Prior to installation in the EBR-I, Mark IV core, the fuel rods may be handled manually for short periods for purposes of inspection, inventory, and loading. After irradiation in the core, all fuel must be

handled by remote techniques. All handling procedures occurring both before and after use in the reactor are thoroughly monitored by techniques noted in the main body of this report.

As of the date of preparation of this report, EBR-I, Mark IV, was in routine operation. To date no measurable plutonium aerosols have been produced nor has any escape of plutonium from the reactor or from individual fuel rods been detected.

C. EBR-II

1. Reactor. The Experimental Breeder Reactor II is a fast, unmoderated, sodium-cooled power breeder reactor, with a design power rating of 62.5 Mwt. Electric generating capacity is 20 Mwe. EBR-II was conceived as an integrated power plant and fuel-reprocessing facility. A pyrometallurgical processing plant will receive the spent fuel, remove certain fission products, and reform the purified fuel into rods and subassemblies to be returned to the reactor for further use.

2. Fuel. The first core of EBR-II uses an alloy of 50% enriched uranium plus 5 w/o synthetic fission products. A second core has been proposed for EBR-II, to consist of plutonium as the fissile material alloyed with uranium and synthetic fission products. One proposed alloy is: U-20 w/o Pu-10 w/o synthetic fission products. Individual fuel rods would be stainless steel tubes containing slugs of fuel and fertile material, and filled with sodium for thermal bonding. It is reported that the addition of the synthetic fission products (used to avoid large changes in alloy composition with each fuel cycle) reduces or eliminates the pyrophoricity problem.(18)

D. The Fast Reactor Test Facility (FARET)

1. Reactor. The Fast Reactor Test Facility (FARET) is a fast reactor facility tentatively proposed for construction at the EBR-II area of the NRTS. FARET is an experimental facility for (1) high-temperature and high power-density performance tests of advanced fast reactor cores and (2) physics experiments for the evaluation of the neutronics and dynamics of core systems. FARET will operate at power levels up to 50 Mwt and at core temperatures up to 1,200°F (650°C). The coolant is sodium.

2. Fuel. The fuels proposed for use in FARET are primarily ceramics, such as the oxides, carbides, sulfides, and nitrides of uranium and plutonium. The initial loading is scheduled to consist of a uranium ceramic. Subsequent loadings will contain plutonium. The physical and radiological characteristics of the aerosols of these materials are largely unknown.

E. The Zero Power Plutonium Reactor (ZPPR)

1. Reactor. The Zero Power Plutonium Reactor (ZPPR) is a facility proposed for construction by the Argonne National Laboratory at the EBR-II area of the NRTS. The purpose of ZPPR is the study of the reactor physics of large plutonium-fueled fast power reactors. ZPPR is essentially an advanced, enlarged version of ZPR-III. All building facilities are designed for the use of plutonium fuels.

2. Fuel. ZPPR has an extremely flexible fuel-loading capability. Fuel assemblies will consist of metal drawers containing plates or slugs of fuel and other materials used in the construction of reactors. Fuels to be studied will include plutonium metal alloys, and PuO_2 and PuC ceramics. All plutonium fuel plates or slugs will be jacketed to reduce contamination to a minimum.

APPENDIX C

DIAGRAMS AND PHOTOGRAPHS OF SAMPLING AND MONITORING DEVICES

The diagrams and photographs in this Appendix are concerned with the locally originated or modified units discussed in the main body of this report.

Ratio Air Monitor

The following diagrams show local modifications of the ratio monitor described in Section D, 1, of this report.

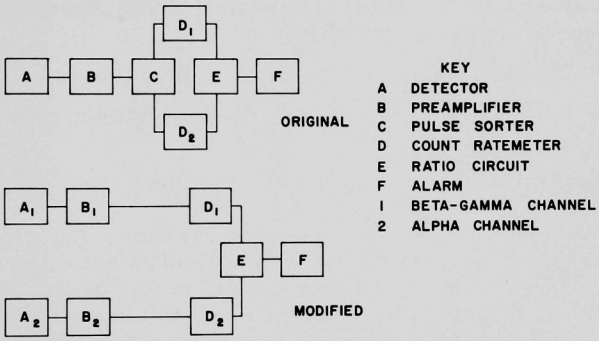

Fig. C-1. Block Diagram of Modified Ratio Air Monitor

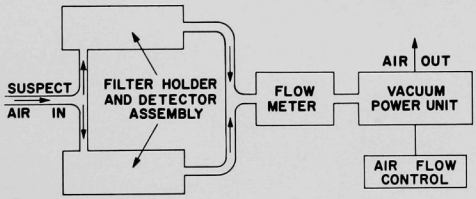

Fig. C-2. Block Diagram of Air Flow System, Modified Ratio Air Monitor

AK-1 Continuous Alpha Air Monitor

The AK-1 is a continuously operating, alpha-detecting, particulate air monitor of conventional design. It was conceived as a high-level monitor for use in plutonium operations in the vicinity of operating nuclear reactors. Design considerations included the following items:

a. Unit should be relatively insensitive to variations of the ambient gamma-radiation background and to accumulation of beta-gamma emitting particulates.

b. Unit should have a stable predictable response and long-term reliability. False alarms should be minimal.

c. Unit should be capable of operating unattended for extended periods of time.

d. Unit should sample room air at a rate of not less than 15 m^3/hr.

e. Unit should use conventional, commercially available components wherever possible.

f. Unit should be constructed at a minimum cost.

The basic unit consists of a sampling system, collecting aerosols on HV-70 filter paper at a rate of 0.25 m^3/min, and a detection system, composed of a ten-wire, gas-flow proportional probe, feeding into a sensitive preamplifier and a compact count-rate meter. An alarm is provided by a relay meter in the count-rate-meter circuit and an electric buzzer. Provision is made for remote alarm. Output from the count-rate meter is recorded on a strip-chart recorder. Control systems are provided for control of air flow rate and detector gas flow.

The entire system is cart mounted and is wheel-portable. The unit shown in the photographs was subsequently modified by mounting the counting gas cylinder on the cart for full mobility.

The unit has been very stable in operation and all components have been satisfactory under long term continuous use. Carbon brushes on the vacuum power unit must be replaced at approximately 90-day intervals.

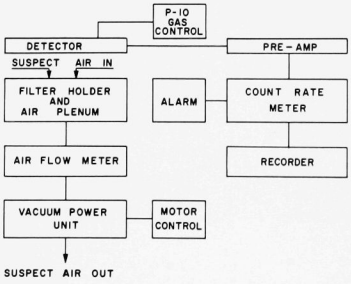

Fig. C-3

Block Diagram of Continuous Alpha Air Monitor - High Level - Model AK-1

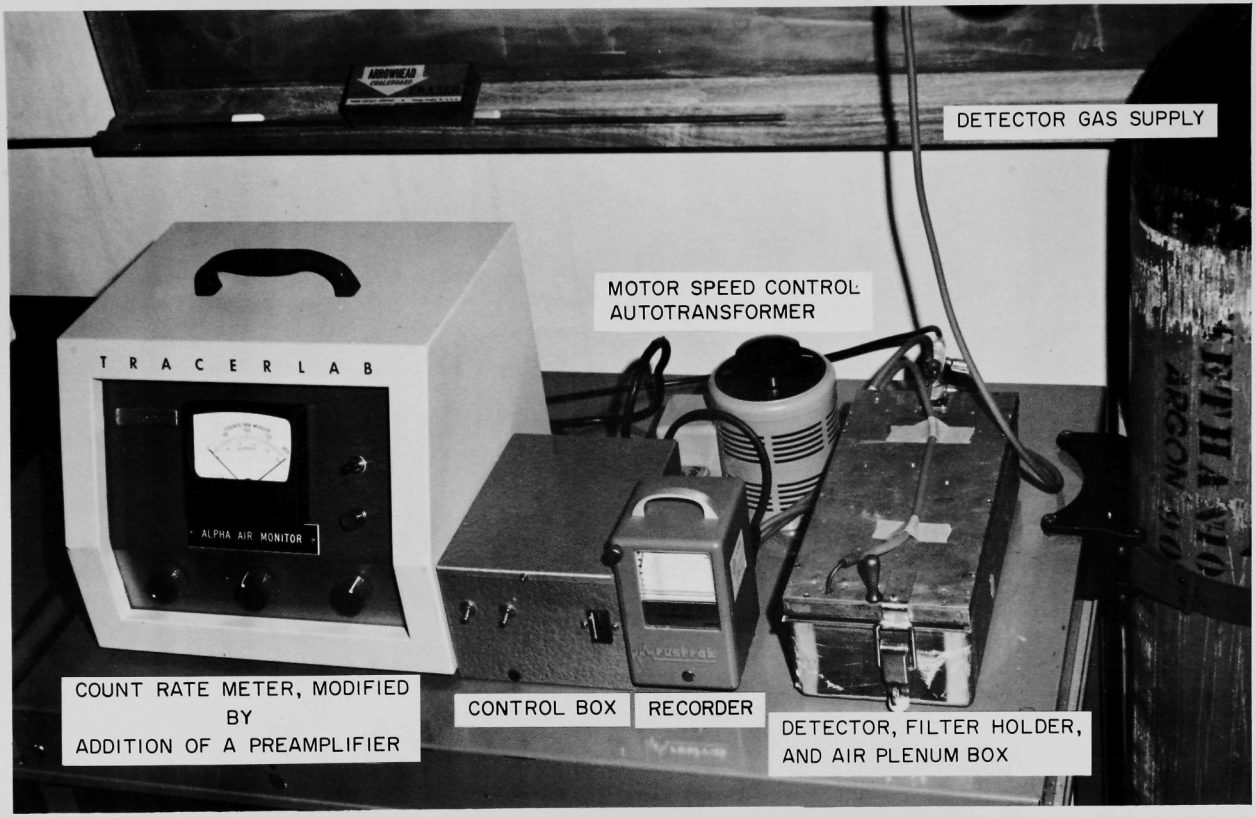
DETECTOR GAS SUPPLY
MOTOR SPEED CONTROL
AUTOTRANSFORMER
TRACERLAB
ALPHA AIR MONITOR
COUNT RATE METER, MODIFIED
BY
ADDITION OF A PREAMPLIFIER
CONTROL BOX
RECORDER
DETECTOR, FILTER HOLDER,
AND AIR PLENUM BOX

Fig. C-4

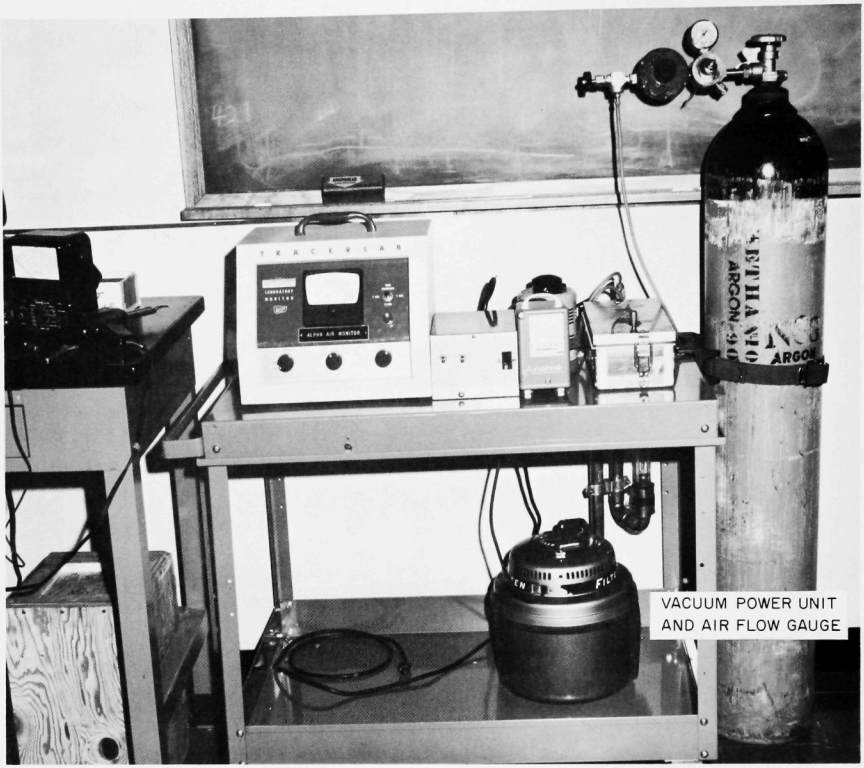

Fig. C-5

"SNIFFER" Alpha Air Monitor

The "SNIFFER" was designed for the rapid detection of alpha-emitting particulate aerosols in areas or places where it would be impossible or impractical to use conventional air monitors. The original application was the monitoring of crevices and openings in plutonium-transfer casks, but this has been extended to include the monitoring of small inaccessible areas, ducts, and openings where contamination of air by plutonium is suspected.

The unit is basically a portable air sampler coupled to an Eberline PAC-3G portable alpha monitor. Air is drawn through a "snout" consisting of a length of metal tubing, and is deposited on a 2 x 8-in. HV-70 filter. A gas-flow, alpha-sensitive detector is mounted above the filter paper collection surface. Calibrated efficiency of the detector is

approximately 45% for plutonium alphas. A Pu^{239}-alpha calibration source is built into the detection-filter assembly. A flexible hose was used to connect the vacuum power unit to the sampler for mobility.

The "SNIFFER" is designed as a "go or no go" device for qualitative identification or detection of airborne alpha activity. In an atmosphere containing 4,400 dis/min-m^3 [1,000 x MPC(40) for plutonium] of an alpha emitter, the filter will accumulate approximately 1,100 dis/min (alpha) per minute of sampling. At a detector efficiency of approximately 45%, the meter would indicate 490 to 500 cpm at the end of the first minute of sampling. For greater or lesser concentrations, response is proportional to relative concentration.

Radon-thoron daughters will accumulate on the filter and must be discounted when evaluating the meter readout.

Fig. C-6. "SNIFFER" - A Portable Unit for Detection of Alpha-emitting Aerosols

APPENDIX D

GLOSSARY

Some of the terms used in this report are based on local or ANL usage and may not conform to terms used elsewhere. The following definitions are offered in the interest of subject clarity.

AEROSOL is a term which may be applied to any dispersion of solid or liquid particles of microscopic size in gaseous media. Plutonium aerosols, in the sense used in this report, are dispersions of solid particles of plutonium or plutonium compounds in air.

AIR SAMPLING is the process of collecting samples of air by any of several methods. The samples collected are analyzed or counted at some location separate from the sampling unit.

AIR MONITORING is the process of simultaneously collecting and counting an air sample. The air monitors described in this report are self-contained units, i.e., sample collection, detection, counting, and recording are all provided as functions of a single unit.

ANL-IDAHO denotes the Argonne National Laboratory's Idaho Facilities, located on the National Reactor Testing Station in Idaho.

CONSTANT monitoring or sampling devices are operated on an essentially continuous basis. Spot or grab sampling or monitoring implies short-term operation, i.e., periods ranging from seconds up to several hours.

LOW LEVELS of plutonium aerosols are considered to be those concentrations ranging from zero to 100% of MPC(40). High levels of plutonium aerosols are considered to be those concentrations in excess of 100% of MPC(40).

NRTS is the abbreviation of the National Reactor Testing Station. The NRTS is located west of the city of Idaho Falls, Idaho.

Ra-DEF refers to a mixture of radioactive material, used as a "secondary" counting standard. It is an equilibrium radium decay-product material consisting of Radium-D (Pb^{210}), Radium-E (Bi^{210}), and Radium-F (Po^{210}).

THORON (Tn) is a name frequently applied to the isotope $_{86}Rn^{220}$. Its precursors are $_{88}Ra^{224}$ (Thorium-X) and $_{90}Th^{228}$ (RdTh). It is used in this report to imply its origin from thorium.